Scintillation Method of Analysis for Determination of Properties of Wear Particles in Lubricating Oils

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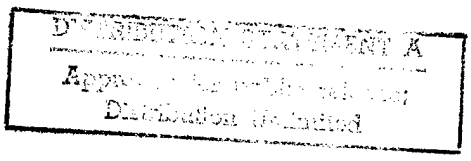

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Abstract: Nowaday there is a demand for effective methods and equipment for early detection of failures of aircraft engines. The spectral diagnostics methods used in Russian civil aviation have a low information capacity and poor metrological features and they can not be used for reliable prognosis of the engine serviceability. Applied Physics Institute of Irkutsk State University (Russia) together with Joint-Stock Company «Baikal Airlines» have elaborated the scintillation method of analysis of lubricating oils for wear products. It enables to obtain quickly and with high accuracy the information on

- a metal wear particles content,
- a dissolved metal content,
- an amount of wear particles,
- an amount of simple particles,
- an amount of complex particles,
- a composition of each particle.

The exact determination of wear particle composition (including micron-sized particles of Fe-Cu, Fe-Ni, Fe-Ag) permits to pursue the unit-to-unit diagnostics of aircraft engines.
In addition to the civil aviation the scintillation method is applicable for diagnostics of an equipments in airforce, a navy, a petroleum and engineering industries, in automobile, railway and sea transport and other engines and mashines. Besides, this method may be useful for tribological investigation of a quality of lubricant materials, in the development of new lubricants.

Key words: Aircraft engine; ferrograghic analysis; graduation; lubricating oils; metrological properties; scintillation spectral analysis; spectrometer; unit-to-unit diagnostics; wear particles.

Introduction: In accordance to data of Rolls-Royce Company 50 per cents of mechanical faults of aircraft engines are being detected with the use of tribological methods [1]. By the tribodiagnostics is meant techniques of continuous measurements of a content and a number of ferromagnetic particles with the use of on-line sensors and, in addition, the methods of periodical off-line measurements. However the sensors can come into false action and incorrect conclusions may be done on oil fitness. For instance, the sensors ODDS (in accordance to Tedeco Vickers) give 30 % of false actions.

The method of periodical laboratory checks are more time consuming, but they enable to predict exactly the work features of an engine.

Among the laboratory methods the ferrographic and emission spectral techniques can be called first of all, which provide a great body of data on wear particle. Now a loss of interest in emission spectral methods should be noted. It is associated with that the serviceability of an engine can be judged from the value of content of wear particles. Since the element content is measured with a low accuracy and depends on many random parameters, the efficacy of this method is insufficient [2]. Nevertheless so far emission spectrometers have being produced by the firm Baird for Defense Department of USA [3], and emission spectral method is one of the main techniques for failure prediction in Russia.
Ferrographic method gives the maximum of information on wear products- the size of particles, its shape, the metal content and the alloy type. But it is very time consuming and in many cases does not give the opportunity of material identification. A low resolution in process of alloy determination (for instance, iron alloy with a different content of chrome) limits the possibilities of unit-to-unit diagnostics. The problem is also the correct determination of admissible values of wear particles features.

From the above it is clear that the modern diagnostics equipment has a number of disadvantages reducing the efficacy of its use. Some of them (determination of a particle size by on-line sensors) are of fundamental nature and can not be eliminated in the frames of these methods.

The simultaneous application of the ferrographic and emission spectral methods can give the required information on wear products in oil. Such set of equipment is used in the service center of the airport «Sheremetievo». But the price of this equipment is about $400 000 and small aircraft companies can not afford to buy it. Hence the elaboration of new not expensive methods providing maximum of information about wear particles is vital now.

One of such methods is a scintillation method of analysis [4] based on serial inputting of wear particles in the area of spectrum excitation and detection of radiation signals from these particles. Let us remember the principle of its operation. (the figure 1).

The principle of scintillation spectrometer operation: The preliminary prepared sample is spraying by the special ultra-sonic sprayer (1). Obtained spray consisting of oil droplets and wear metal particles is blowing in plasma of a UHF-discharge (2) by the gas stream. The temperature of plasma is 5200 K. The sprayer works so that wear particles resided in oils go into plasma sequentially one by one. When a particle fell in plasma it is heated, evaporated and the atomic vapor obtained is excited. A flash (a scintillation) of a particle occurs.

A condenser (3) focuses the radiation onto a spectral device (4). The polychromator (4) resolves the radiation spectrum that is detected by the photomultipliers (5-7). The duration of a radiation pulse of a particle is proportional to the duration of its stay on plasma and comprises 1-10 ms, the amplitude (the area of a pulse) is proportional to the evaporated mass of a particle. Thus there is a sequence of pulses with different amplitude and a

different duration on the output of photomultipliers. Electrical pulses come from them into the analog-digital converter (8) and are treated by the computer. The pulse signal corresponds to wear particles, a continuous signal is bound with dissolved metal.

The figure 1 shows only three channels to pick out the signals. Its number depends on a type of polychromator and may be increased. Each channel is tuned to the detection of the spectral lines of given element.

As simple particle, consisting only one element, enter the plasma (for example, iron particle), a sequence of radiation pulses is present only at one channel (the channel 5 in the figure 1(b)). At the channels 6 and 7 a continuous weak background radiation of plasma is observed.
In the case when complex metal particles, consisting of some elements and simple particles reside simultaneously in oil the computer analyzed the times of a signals appearance.

If two or more pulses agree in time it points to a complex composition of the particle. In the figure 1 (b) pulses coincident with each other in time at the channels for Fe and Mn are shown.

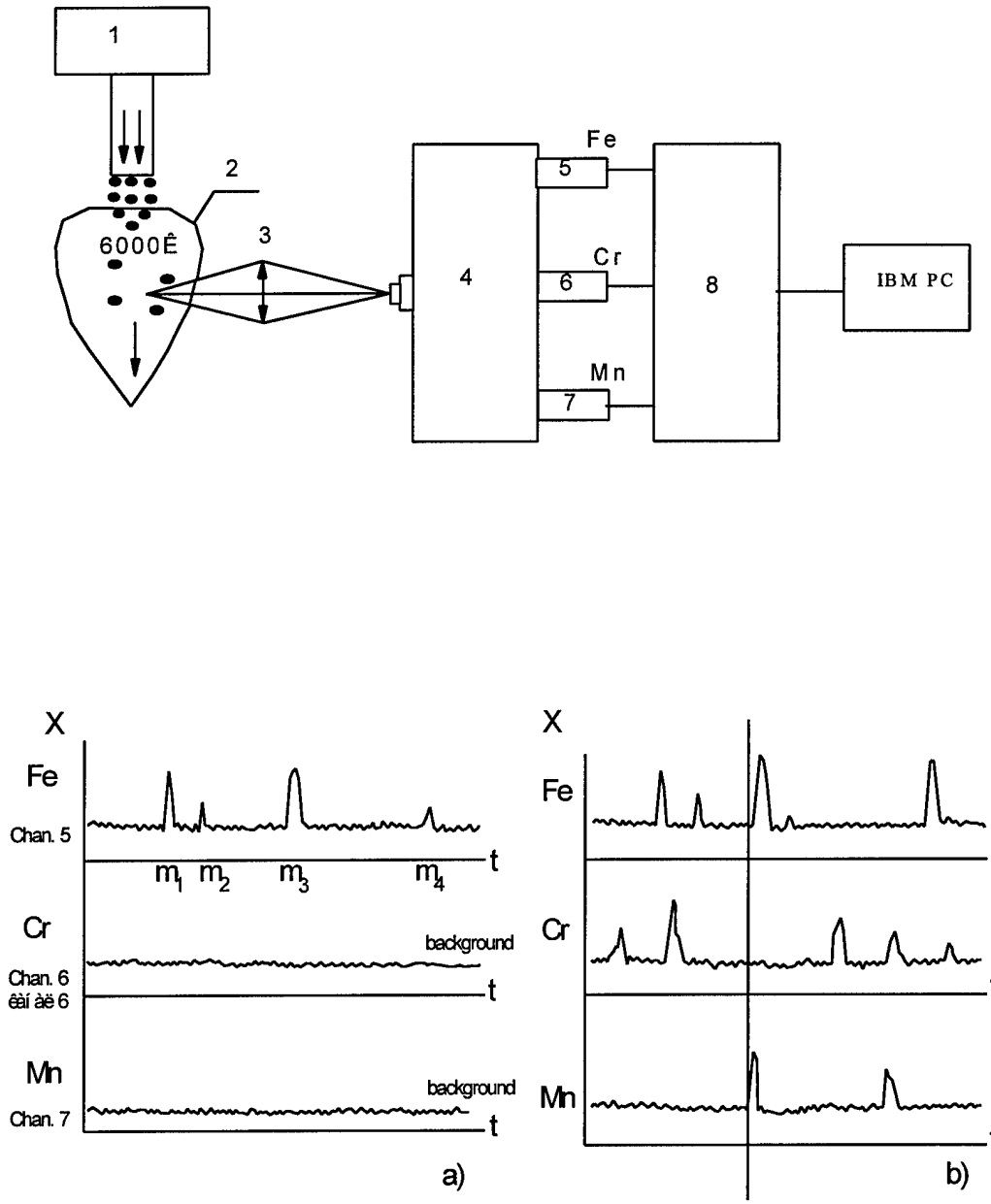

Figure 1. The sketch of scintillation spectrometer for three channels. The sequence of radiation pulses a) for one element being determined, b) for three elements

The standard deviation for determination of a stechiometric composition S was evaluated on reference particles, specially prepared, with known composition. It was less than 0.25. Hence it follows that dependence of scintillation signal on particle path does not play a decisive role.

Results on particle composition were compared with those obtained on microanalyzer Camebax SX-50. They agree very closely.

Thus from the above it is seen that the scintillation method provides the possibility within 5 minutes to obtain the information about:

- a metal wear particles content,
- a dissolved metal content,
- an amount of wear particles,
- an amount of simple particles,
- an amount of complex particles,
- a composition of each particle.

It should be added that stabilization of a discharge in UHF plasma generator the air was used supplied through the special filter. The use of air reduces the metrological properties of the method, but it saves the operators the trouble and expense of using argon and nitrogen. The discharge chamber of plasma generator can work without failures during 1000 hours.

The graduation of the scintillation spectrometer when both continuous and pulse signals are detected: For graduation of scintillation spectrometer the following system of equation are used:

$$\begin{aligned} \overline{F} &= a_1 + a_2\left(C_L + C_P^1\right) \\ B &= b_2 \cdot C_P^2 \\ C_P &= C_P^1 + C_P^2 \end{aligned} \qquad (1)$$

where $\overline{F}$ - is continuous signal, B- is a pulse signal, $\tilde{N}^1_{Đ}$ is the content submicron-sized impurities, $\tilde{N}^2_{Đ}$ is the content of large particles, $\tilde{N}_L$ is the content of dissolved metal.
We assume here that:

- dissolved metal does not provide scintillation pulses
- submicron-sized particles, for which the scintillation principle breaks down (that is the analytical area has more than one particle), gives rise to the spectrum background, similar to dissolved metal,
- large pulses do not effect a background component of a signal, and a small pulses do not give a contribution in the pulse component. Thus a content of wear particles is split into two components $\tilde{N}^1_{Đ}$ and $\tilde{N}^2_{Đ}$, the sum of which results in the total metal content.

To obtain the graduation curve the three coefficients must be defined: a_1, a_2 b_2. It may be done in two stages:

1. To find the parameters a_1 and a_2 the standard samples are used, which have only dissolved metal (the standard Conostan). In this case $\tilde{N}^1_{Đ}$ =0 and graduation procedure involves the solution of the equation
$$\overline{F} = a_1 + a_2 \cdot C_L \qquad (2)$$
2. To find the parameters $\tilde{N}^1_{Đ}$ and $\tilde{N}^2_{Đ}$ the standard samples are used, which have only discrete impurities. From eq.(2) the content of submicron-sized particles is determined as
$$C_P^1 = \frac{\overline{F} - a_1}{a_2} \qquad (3)$$

and the content of metal in large particles

$$C_P^2 = C_P - \frac{\overline{F} - a_1}{a_2} \quad (4)$$

After this the regression dependence for analytical parameters B is constructed as:

$$B = b_2 \cdot \left(C_P - \frac{\overline{F} - a_1}{a_2} \right) \quad (5)$$

The limit of detection and reproducibility of the method: The apparatus had two channels tuned on the spectral lines FeI 302.0 nm and CuI 324.3 nm. The flashes of these spectral lines were detected. In doing so, we detected a number of scintillation pulses N (a number of particles detected) and the total area S of pulses was measured. The both values were used as the parameters associated with the content of elements in oil.

The goal of the experiment was the evaluation of accuracy and limits of detection of metal in oil. For this purpose the reference samples were prepared where wear particles were simulated by the oxide power Fe_2O_3 and Cu_2O [5]. As the base the oils MGD-D, MS-8P, SM-4.5 and B3-V were taken.

The use of different oil bases enables to evaluate the influence of oil type on a signal level. As analytical parameter the average X over 20 separate measurements, with excluding the gross errors, was taken.

The graduation curve has been constructed for all types of oil (the table I).

Table I

The content of Cu and Fe in "clean" oils C_{idle}

Type of oil	The content of metal C_{idle}, μg/g	
	Cu	Fe
MGD-D	0,4	0,9
MS-8P	0,3	1,4
SM-4.5	0,1	0,3
B3-V	0,1	0,7

The confidence interval $\Delta C = \pm 2\sigma_{\Sigma}/C$ in working span of contents was calculated for a single measurement at reliability P=0.95 in accordance with [2]. The value of standard deviation σ_C of a single measurement was gained from the standard deviation of separate measurements of signal level on graduation plot $\sigma_{\tilde{O}}$. The value ΔC for all the types of oils was approximately the same. The table II lists the averaged values for ΔC.

Table II

The confidence interval for single measurement of metals content by the scintillation method

Cu	C,μg/g	1	2	3	4	5	
	ΔC, (%)	17	14	12	16	17	ΔC, (%)=15
Fe	C,μg/g	2	4	6	8	10	
	ΔC, (%)	30	30	35	30	35	ΔC, (%)=32

This value remains practically constant with a change of the metal content. It provides reason to take the mean value ΔC as a constant in whole range of contents being determined and for all oils.

The value m_{min} may be thought of as limit of detection in working spans of contents (the table III), according to [6].

Table III

The value m_{min} for the scintillation method of analysis

Oil		Cu			Fe		
Oil	Parameter	C_{idle}, μg/g	V_{const}	m_{min}, μg/g	C_{idle}, μg/g	V_{const}	m_{min}, μg/g
MGD-D (O1)	N		0,06	0,1		0,13	0,7
		0.4			0.9		
	S		0.06	0.1		**	-
MS-8P(O2)	N		0,03	0,04		0,11	0,8
		0.3			1.4		
	S		0.03	0.04		**	-
CM-4.5(O3)	N		0,04	0,02		0,14	0,3
		0.1			0.3		
	S		0.06	0.03		0.14	0.3
B3-V(O4)	N		0,06	0,03		0,08	0,3
		0.1			0.7		
	S		0.06	0.03		0.13	0.5

The average from O1 to O4: $\overline{m}_{\min Cu} = 0.05$ μg/g, $\overline{m}_{\min Fe} = 0.5$ μg/g.

Thus, the accuracy of the scintillation method complies fully with requirements in [5]. Results have been obtained on the laboratory model of the scintillation spectrometer. After advancement of the apparatus and instructional materials the accuracy may be notably enhanced.

Unit-to-unit diagnostics of aircraft engines on scintillation measurements: To localize the failure the data from the bill of parts and units of aircraft engines had been used. The bill listed the information on parts and units working in oil, a mark of material and chemical composition of material.

Table IV gives indications that have been obtained from chemical composition of materials and combined on units. As it is seen from the table IV the engine is divided into 7 units.

The name of wear indication, for example Fe (Cr11Ni10), points to basis element of alloy, alloying elements and their percentage are given in brackets. In doing so, the alloying elements are taken into account, the content of which is sufficiently large and detection of which in wear products is most probable.

The indication on wear hardening particles is listed separately. Such particles may arise by interaction of engine parts with the different composition or different composition of coating. The presence in oil of wears particles of definite wear indication or the group of wears indication points to wear of corresponding parts and units.

Thus, the presence of particles of Fe(W9), Cu(Zn34), Al(Cu5), exceeding some limit values in the content and in the number, is indicative of an intensive wear of bearings, and particles of Ti and Mg(Al9) testifies to a failure of a starter.

The situation is possible when particles Fe(Cr12) are detected, which may belong to may units of engine. In this case the particles of hardenings should be taken into account. If, for instance, together with Fe(Cr12) there are particles TiAl then more likely the defective unit as is a compressor.

Table IV

Characteristic indications of engine parts wear

On material composition							
	Engine units						
Material	Starter	Bearings	Transmission	Compressor	Separator body and drives body	Turbine	Oil units
Fe	o*	o*	o*	*	*		*
Fe(Cr12)	*		*	*	*	*	*
Fe(Cr19)					*		
Fe(Cr11 Ni10)	*		*		*		
Fe(W9)		o*					
Cu(Zn34)		*					
Cu(Zn6)	*						
Cu(Pb27)							*
Cu(Sn7)	*						
Cu(Al11)		o*					
Al				*			
Al(Cu5)		o*					
Mg							*
Mg(Al 9)	*						
Ti	o*						
Ag		*					
On wear hardenings							
	(FeCu) (FeMg) (FeTi)	(FeCu) (FeAg) (CuAg) (FeAl)		(FeAl) (TiAl) (FeTi)			(FeMg) (FeCu)

The table V gives the results of scintillation measurements for wear particles in oil of an intact engine with the working time 3092 hours.

Table V

The results of scintillation measurements of properties of wear particles in oil of an intact engine. The time of its work is 3092 hours.

The work time is 3092. Oil Turbonicol	**Fe**					**Cu**				
	N	**$C_d+\tilde{N}_p$, μg/g**	**D, con**	**$\tilde{N}_{atk}$**	**$\tilde{N}_{aa}$**	**N**	**$C_d+\tilde{N}_p$, μg/g**	**D, conv**	**$\tilde{N}_{atk}$**	**$\tilde{N}_{aa}$**
	4	0 + 0.02	1.10	0.2	-	6	0.0+<0.01	4.0	0.1	-
	Number of complex particles					Number of complex particles				
	FeMg 0	FeCu 0	FeAg 0	FeNi 0	FeAl 0	CuMg 0	CuFe 0	CuAg 0	CuAl 0	CuNi 0
The pouring oil after the ferrograph study	6	0.02+0.16	1.2	-	-	37	0.0+0.01	3.2	-	-
	Number of complex particles					Number of complex particles				
	FeMg 2	FeCu 0	FeAg 0	FeNi 0	FeAl 0	CuMg 9	CuFe 0	CuAg 0	CuAl 0	CuNi 0

Ag					**Al**				
N	**$C_d+\tilde{N}_p$, μg/g**	**D, conv**	**$\tilde{N}_{atk}$**	**$\tilde{N}_{aa}$**	**N**	**$C_d+\tilde{N}_p$, μg/g**	**D, conv**	**$\tilde{N}_{atk}$**	**$\tilde{N}_{aa}$**
1	0.0+0.01	3.0	-	-	0	0	0	-	-
Number of complex particles					Number of complex particles				
AgFe 0	AgCu 0	AgAl 0	AgMg 0	AgNi 0	AlFe 0	AlCu 0	AlAg 0	AlNi 0	AlMg 0
0	0.0+0.0	0	-	-	2	0 + 0.01	0.15	-	-
Number of complex particles					Number of complex particles				
AgFe 0	AgCu 0	AgAl 0	AgMg 0	AgNi 0	AlFe 0	AlCu 0	AlAg 0	AlNi 0	AlMg 0

Ni					**Mg**				
N	**$C_d+\tilde{N}_p$, μg/g**	**D, conv**	**$\tilde{N}_{atk}$**	**$\tilde{N}_{aa}$**	**N**	**$C_d+\tilde{N}_p$, μg/g**	**D, conv**	**$\tilde{N}_{atk}$**	**$\tilde{N}_{aa}$**
0	0	0	-	-	23	0.00+0.01	0.8	-	-
Number of complex particles					Number of complex particles				
NiFe 0	NiCu 0	NiAg 0	NiAl 0	NiMg 0	MgFe 0	MgCu 0	MgAg 0	MgAl 0	MgNi 0
0	0.0+0.0	0	-	-	80	0.0+0.04	4.0	-	-
Number of complex particles					Number of complex particles				
NiFe 0	NiCu 0	NiAg 0	NiAl 0	NiMg 0	MgFe 2	MgCu 9	MgAg 0	MgAl 0	MgNi 0

Here: *N* is a number of pulses detected, which is proportional to a number of wear particles, C_d is the element content of dissolved metal and submicron-sized particles, C_p is

the element content of wear particles, D_{conv} is the parameter proportional to the mean size of particles, C_{aa} and C_{atk} are the total content of metal determined by the different methods.

Noteworthy is here a small number of detected particles of all elements, a low content of wear particles and almost full absence of complex particles.

The table VI contains the results of ferrographic and emission spectral analysis of the same engine, made in technical center of airport "Sheremetevo". Analysis did not reveal any disrepair in its work.

Table VI

The results of ferrographic and emission spectral analysis of the same engine on the spectrometer MOA

Type of particles	**Size**	**Missing**	**a little amount**	**a mean amount**	**a large amount**
Normal rubbing wear	<15 μm		x		
Fatigue crumbling	>20 μm		x		
Spherical particles	to 10 μm	x			
Plate particles	>15 μm	x			
Particles of hard wear	>15μm		x		
Particles of cutting wear	to 20 μm		x		
Particles of corrosion wear		x			
Particles of oxides		x			
Ferrous particles			x		
Nonferrous particles		x			
Nonmetal particles			x		
Nonmetal particles (amorphous)		x			

Conclusion on the wear of engine				
Weak	Normal	Warning	Intensive /red signal	
	Õ			

Emission spectrometer MOA , μg/g

Al	**Fe**	**Ag**	**Cu**	**Mg**	**Ti**	**Ni**	**Si**
00.0	0.2	0.0	0.1	0.0	0.0	0.0	0.0

In the table V the data of scintillation measurements are given for oils which has been collected and analyzed after ferrographic study, including washing of glass plate by the detergent. These results are given in the line "pouring oils".

On the whole, the pouring oils obtained after the ferrographic measurements have been analyzed for 12 engines. The increased number of detected particles in relation to the

source samples has been obtained. The mean size of particles in pouring oils turns to be less.

On of the explanation consists in the following.

At micro-roentgen study of the wear particles on the analyzer Camebax SX-50 the particles of resinous deposits with metal impurities were detected. On addition of the detergent these resinous particles were dissolved, resulting in the increase of a number of particles in oil sample. The fact revealed is of fundamental practical importance.

It is seen from the table V that the pouring oils contain rather large amount of particles, including ferromagnetic ones. It has been found that a number of particles in pouring oils varies random from sample to sample. This fact can lead to an essential distortion of the true values of wear parameters, and, hence, the evolving failure can be missed.
On the other hand, the results of scintillation measurements can be used as an additional diagnostics parameters, since the resinous substances with metal particles testify the quality of oil.

The table VII gives the results of scintillation measurements for the engine taken from the service.

Table VII

The results of scintillation measurements for an engine taken from the service. The work time is 703 hours

The work time is 703 hours. Oil Turbonicol	Al					Ni				
	N	$C_d+\tilde{N}_p$, μg/g	D, conv	$\tilde{N}_{atk}$	$\tilde{N}_{aa}$	N	$C_d+\tilde{N}_p$, μg/g	D, conv	$\tilde{N}_{atk}$	$\tilde{N}_{aa}$
	11	0.0+0.5	0.06	-	-	450	0.21+0.3	0.3	-	-
	Number of complex particles					Number of complex particles				
	AlFe	AlCu	AlAg	AlNi	AlMg	NiFe	NiCu	NiAg	NiAl	NiMg
	2	1	0	0	1	449	0	0	0	0

Mg					Ag				
N	$C_d+\tilde{N}_p$, μg/g	D, conv	$\tilde{N}_{atk}$	$\tilde{N}_{aa}$	N	$C_d+\tilde{N}_p$, μg/g	D, conv	$\tilde{N}_{atk}$	$\tilde{N}_{aa}$
640	0.30+0.09	0.42	-	-	226	0.04+0.52	1.05	-	-
Number of complex particles					Number of complex particles				
MgFe	MgCu	MgAg	MgAl	MgNi	AgFe	AgCu	AgAl	AgMg	AgNi
24	23	4	1	0	9	0	0	4	0

Fe					Cu				
N	$C_d+\tilde{N}_p$, μg/g	D, conv	$\tilde{N}_{atk}$	$\tilde{N}_{aa}$	N	$C_d+\tilde{N}_p$, μg/g	D, conv	$\tilde{N}_{atk}$	$\tilde{N}_{aa}$
1640	5.3+2.7	0.25	-	-	520	0.35+0.2	2.0	-	-
Number of complex particles					Number of complex particles				
FeMg	FeCu	FeAg	FeNi	FeAl	CuMg	CuFe	CuAg	CuAl	CuNi
24	20	9	449	0	23	20	1	1	5

Unfortunately, the table does not contain data on Cr and Ti. These elements were not allowed by the used polychromator.

The comparison of the tables V and VII shows that a number of simple particles (exclusive of Al) and a number of complex ones has increased significantly. From the alloys only FeNi has been found.

From the table IV it is seen that FeNi can belong to a starter, to details of a drives body and of a separator body.

Additional indications on hardenings of latest two units are missing. Hence the hardenings FeCu and FeMg points to the fault in the starter. Besides, the increase of a number of FeAg particles testifies on the initial stage of a wear of intershaft bearing.
The taking the engine to pieces has confirmed the results made.

Conclusions: 1. The laboratory model of the scintillation spectrometer for a spectral analysis of liquids for wear products has been elaborated.

As the spectral light source the elaborated UHF plasma generator of a cyclone type was used. It is rather reliable in operation (guaranteed is 1000 hours of work), the air may be used as a base gas supplied from the compressor.

2. It enables for time 5 minutes to obtain the information on
- a metal wear particles content,
- a dissolved metal content,
- an amount of wear particles,
- an amount of simple particles,
- an amount of complex particles,
- a composition of each particle.

The procedure of spectrometer graduation on dissolved metal and metal particles has been elaborated. The metrological properties of the method have been evaluated. It was shown that the accuracy of the method complies fully with the customary requirements.

2. The emission study enables unit-to-unit diagnostics.
Results have been obtained on the laboratory model of the scintillation spectrometer. After an advancement of the registration apparatus and instructional materials an accuracy of the method may be notably enhanced.

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